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### BIOREMEDIATION OF PETROLEUM HYDROCARBON IN CONTAMINATED SOILS: COMPARISON OF COMPOST AND WWTP SLUDGE RESIDUAL ADDITION

### BIOREMEDIASI TANAH TERCEMAR PETROLEUM HYDROCARBON: PERBANDINGAN PENAMBAHAN KOMPOS DAN LUMPUR IPAL

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#### ABSTRAK

Kegiatan pengelolaan minyak bumi terus meningkat, maka dari itu dibutuhkan tindakan penanganan pemulihan kondisi lingkungan yang disebabkan oleh kegiatan tersebut. Penelitian ini bertujuan untuk mengetahui perbedaan laju pertumbuhan bakteri dan efisiensi penyisihan Total Petroleum Hydrocarbon (TPH) dengan variasi perlakuan pemberian kompos dan lumpur residu pengelolaan air limbah. Penelitian dilakukan dalam skala laboratorium dengan konsentrasi awal TPH sebesar 5,5% selama 5 minggu atau sampai TPH mencapai konsentrasi kurang dari 1% sesuai dengan baku mutu. Tanah yang digunakan berasal dari Pantai Marunda, Bekasi. Kompos berasal dari UPS (Unit Pengolah Sampah) Merdeka, Depok. Lumpur Instalasi Pengelolaan Air Limbah (IPAL) berasal dari Jababeka. Isolat bakteri yang digunakan berasal dari tanah tercemar TPH disekitar ,kilang minyak. Hasil dari penelitian menunjukkan laju pertumbuhan bakteri pada perlakuan penambahan kompos dan lumpur IPAL pada konsentrasi 5% dan 10% masing-masing adalah 0,7567/minggu dan 1,154/minggu untuk kompos, serta 0,8783/minggu dan 1,1109/minggu untuk residu lumpur IPAL. Efisiensi penyisihan TPH yang diperoleh adalah 95,32% and 96,85% untuk penambahan kompos dan 91,15% dan 91,02% untuk penambahan residu lumpur IPAL pada konsentrasi 5% dan 10%. Berdasarkan hasil uji-t, perbedaan untuk masing-masing perlakuan tidaklah signifikan. Uji korelasi antara perubahan konsentrasi TPH dengan pertumbuhan bakteri menunjukkan hubungan lemah berbanding terbalik.

Kata Kunci: bioremediasi, kompos, lumpur IPAL, penyisihan TPH, laju pertumbuhan bakteri

#### ABSTRACT

Crude oil's processing into energy continue to increase, hence treatment for its environmental impact is needed. The objectives of the study is to determine the differences in bacteria growth rate and removal efficiency of Total Petroleum Hydrocarbon (TPH) between compost and WWTP (Waste Water Treatment Plant) sludge addition at 5% and 10% concentration levels. Those effects were acknowledged through experiments in laboratory scale using soil contaminated by 5,5% TPH within 5 weeks until it reach less than 1% as the requirement. The soil comes from Marunda Beach, compost from UPS Merdeka, WWTP sludge from Jababeka, and bacteria isolated from soil contaminated in the area surrounding refining. The treatment used in this experiment was landfarming with nutrition addition and the main variable analyzed was TPH and the microorganism population. Results of this study show that the bacteria growth rate in

compost and WWTP sludge at 5% and 10% concentration each are 0,7567/weeks and 1,154/week for compost and also 0,8783/week and 1,1109/week for WWTP sludge. The TPH removal efficiency obtained was 95,32% and 96,85% for the addition of compsot as well as 91,15% and 91,02% for the addition of WWTP sludge at 5% and 10% concentrations. Base on a t-Test, the differences between all the variation of concentrations are not significant. The correlation test between TPH degradation to bacteria growth showed that there is a weak downward (negative) linear relationship.

Keywords: bioremediation, landfarming, compost, wwtp sludge, TPH removal, bacteria growth rate

#### I. INTRODUCTION

Petroleum processing activities could lead to soil becoming contaminated by TPH (Total Petroleum Hydrocarbon) (ERM 2009; Sulisyo 2012). In order to treat the contaminated site, bioremediation can be one of the solutions for treatment. Landfarming is one of bioremediation methods that is known as a simple and economical treatment to be applied (Ismail et al. 2013 in Bezza et al. 2015). However, to optimize the treatment, it takes extra nutrients for bacteria to optimally degrade the contaminant (Adams et al. 2015).

Waste Water Treatment Plant (WWTP) sludge residual and compost contains nutrients that can be potentially useful in the bioremediation process. Therefore, it has become essential to investigate the potential utilization of compost and WWTP sludge residual in bioremediation process. This study will determine the differences in the bacterial growth rate and removal efficiency of Total Petroleum Hydrocarbon (TPH) between compost and WWTP sludge addition at 5% and 10% concentration levels.

#### **Theoretical Overview**

Crude oil consists of several hundred chemical compounds that can be described in one term namely Total Petroleum Hydrocarbon (TPH) (ASTDR 1999). Remediation is a strategy to treat land contaminated by various contaminants including in TPH (Suthersan 1999). There are many variables that influence the bioremediation process, such as pollutant characteristics, environmental factors, and indigenous bacteria (Riser-Roberts 1998). Remediation can be classified based on its method: one method is bioremediation. Bioremediation is an environmental treatment that involves microorganisms for detoxification or to degrade the contaminant (Baker et al. 1994). Landfarming is a bioremediation technique that typically utilizes piracy mechanism or stirring the soil, to reduce the level of biological contamination (EPA 2014).

There are many ways to improve landfarming bioremediation processes. For example, addition of more bacteria or more nutrition (Agamunthu P, et al.

2013; Suja et al. 2014). WWTP residual sludge has various nutrition needed by bacteria such as carbon, nitrogen and fosfor (Sarkar et al. 2005). Compost can also add more bioavailability and will strongly affect the development of the microbial community for biodegradation purposes (Chen 2016). Therefore, adding compost or WWTP sludge into the treatment soil can add more bioavailability to the soil and enhance the bioremediation process.

#### **II. METHODOLOGY**

#### A. Material and methods

The study involved experimental research using pots as a reactor and was conducted at the Laboratory of Process, Technology, Research, and Development Center for Oil and Gas Technology (LEMIGAS), Cipulir, Jakarta.

WWTP residual sludge and compost addition for landfarming bioremediation process can increased bacteria population growth and also enhance the biodegradation of contaminant (Agamunthu er al. 2013; Sarkar et al. 2005).

#### **B.** Material Collection

Soil was collected from Marunda Beach, Bekasi. Compost was collected from UPS (Waste Processing Unit) Jalan Merdeka, Depok. Compost was made up 50% kitchen waste and 50% garden waste. WWTP residual sludge was collected from Jababeka Domestic Wastewater Treatment Plant. Compost and WWTP sludge were selected to be added individually into the 5% and 10% (w/w) crude oil-contaminated soil.

#### C. Bacteria Consortia

Three bacteria consortia were used in the treatment. Bacteria were isolated in the laboratory from soil contaminated by crude oil samples collected from the contaminated site. All strains were grown in culture media supplemented with 5% of hydrocarbon content.

#### **D.** Experimental design

The experiment was conducted in a plastic vessel containing 6 Kg crude oil contaminated soil for an

initial TPH concentration of approximately 5200 mg/ Kg. The treatment setup is shown in Table 1 below.

Soil, bacteria consortia, compost or WWTP sludge, and water were mixed thoroughly by a steel hand trowel to achieve a homogeneous distribution of hydrocarbon, water and nutrient. The tanks were tilled every two days with a steel hand trowel.

The soil moisture content was maintained between 13 and 30%. It was preserved by spraying a sufficient volume of deionized water and tilled to ensure uniform oxygen, water and nutrient distribution.

#### E. Parameters and Statistical Analysis

The comparison of bacteria growth and TPH biodegradation in each tank were tested by the t-test method using sigma plot 11.0. The correlation between bacteria growth and the removal of TPH were evaluated by a correlation test using linear regression methods using microsoft excel 2010. Statistical significance was established at the p < 0,05 level.

Table 1Experimental setup				
Tank	Treatment			
A	Addition of 5% compost			
В	Addition of 10% compost			
С	Addition of 5% WWTP sludge			
D	Addition of 10% WWTP sludge			
Control	Control set only for comparison of natural attenuation process			

Table 2
Parameter analysis

1pHSNI 03-6787- 2002Every 3 days2TemperatureSNI 06-6989.23- 2005Every 3 days3TPH ConcentrationUS EPA 9071bEvery 7 days4Microorganism s PopulationBacteriological Analytical ManualEvery 7 days	No	Parameter	Standard Measurement	Interval Time
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<ul> <li>3 TPH US EPA 9071b Every 7 Concentration</li> <li>4 Microorganism Bacteriological severy 7 s Population</li> <li>Analytical days Manual</li> </ul>	2	Temperature	SNI 06-6989.23- 2005	Every 3 days
4 Microorganism <i>Bacteriological</i> Every 7 s Population <i>Analytical</i> days <i>Manual</i>	3	TPH Concentration	US EPA 9071b	Every 7 days
	4	Microorganism s Population	Bacteriological Analytical Manual	Every 7 days

#### **III. RESULTS AND DISCUSSION**

#### A. Bacteria growth curve

The Bacteria growth curves shown in Figures. 1, 2, and 3 explaine the growth rate for each isolated bacteria. It was assumed that the three bacteria need almost three hours for lag phase. For Bacteria A, it takes approximately 9 hours to reach its exponential phase. Bacteria B needs 9 hours to reach exponential phase and to maintain it for the next 1,5 hours. While Bacteria B needs about 7,5 hours to reach the exponential phase. These is different growth curves arise because bacteria have their own growth rate as it is on of their characteristics. The growth rate used is a reference to prepare bacteria consortia to be implemented in the contaminated soil.

#### B. Compost and WWTP Sludge Nutrition Analysis

The nutritional characteristics are exhibited in the table below. WWTP sludge has a higher carbon component but less N, P, and K content.





Table 3 Nutrition analysis of compost and WWTP sludge					
Parameter	Compost	WWTP Sludge			
Moisture content (%)	10,48	78,35			
рН	6,8	6,2			
Carbon (%)	33	34,0			
Nitrogen (%)	0,97	0,47			
Phosphorus (%)	0,76	0,74			
Kalium (%)	1,92	0,08			

The nutritional characteristics are detailed in the table above. WWTP sludge has higher carbon but less in N, P, and K content.

## C. Comparison of bacteria growth rate between compost and WWTP sludge addition

The bacteria growth of contaminated soil throughout the period of the study, (45 days) are shown in Figure 4 for compost amendments and Figure 5 for WWTP sludge addition. The amount of bacteria in soil contaminated and added by compost range between  $5x10^6$  CFU/gr and  $3,3x10^9$  CFU/gr while the contaminated soil added to by WWTP sludge range from  $5x10^6$  CFU/gr and  $1,1 \times 10^{10}$  CFU/gr.

Both of the graphs have a different growth path which might be due to bacteria composition and nutrition ratio. As stated by Jacques Monod, the bacteria growth curve is one of bacteria's physiological characteristics. Therefore, all every bacteria have their own growth curve which is different to others. In addition, the nutrition ratio would also influence the bacteria growth curve. As concluded by Anat Bren (2013), the nutrition ratio is still an issue and because of its complexity it affects bacterial growth.

The bacteria growth rate curve are shown in Figure 3. R<sup>2</sup> determined as bacteria growth rate. The graph shows that treatment with 5% compost addition has a lower bacteria growth rate than treatment with 10% compost addition. It is assumed that the growth rate in 10% compost addition is higher because it has more nutrition than 5% compost addition. As specified by Agamunthu et al. (2013), biostimulation addition will add more bioavailability for microorganisms so it will grow better and





stronger to degrade the contaminant. Moreover as found by Aiyoubi Shahi (2016), bacteria populatinos have increased in the amount of nutrient.

The bacteria growth rate in the treatment with 5% WWTP sludge addition has a higher value than treatment with 10% WWTP sludge addition. It is in opposition to the statement that more bioavailability will lead to a higher bacteria growth rate. Somehow, it can happen through other factors that influence the bacteria growth rate. As identified by Sarles (1956) the bacteria growth rate is influenced by many factors such as food, moisture, temperature, oxygen availability, pH, toxic accumulation, surface

tension, bioavailability and nutrition. Therefore, more nutrition will not always guarantee that the growth rate will be higher. The composition between macro, micro, and trace elements will also influence the bacteria growth rate. As Roshanak et al. (2014) found, in a bacteria community that have more than one species, the population growth had became so



Figure 5 Bacteria growth with WWTP sludge addition during simulation.



TPH concentration and bacteria population during simulation with compost addition.

diverse corresponding to its environmental condition and the composition of its macro, micro nutrient, and trace elements.

At the end of the simulation, two bacteria isolated from land contaminated by TPH were identified as *Staphylococcus hominis* and *Microccus* sp. SK22.

# **D.** Comparison of TPH removal efficiency between compost and WWTP sludge addition

The TPH measured during the simulation (45 days) are shown in Figure.6 for compost addition and Figure 7 for WWTP sludge addition. The graphs show that TPH removal in the treatment with 10% compost addition is higher than the treatment with 5% compost addition. In the first 3 weeks, TPH removal in the treatment with 5% compost addition. It is faster than treatment with 10% compost addition. It is caused by bacteria in the treatment with 5% compost addition having a larger population than treatment with 10% compost addition.

In the 3<sup>rd</sup> week, it can be seen that TPH concentration in the treatment with 10% compost addition decreased faster than treatment with 5% compost addition which coincided with bigger bacteria population growth. It is proof that the carbon from TPH was utilized by bacteria as their carbon source, so that as the bacteria population grew, the TPH removal was decreasing. As stated by Martin Alexander (1994) the decrease in organic content





followed by bacteria growth indicate that the bacteria utilize the nutrition component from the contaminant.

Figure 7 illustrates the change of TPH concentration and bacteria population growth during simulation. During the first weeks, TPH removal in the treatment with 10% WWTP sludge addition was higher than treatment with 5% WWTP sludge addition. This case has shown an inhibitory effect where indigenous bacteria from WWTP sludge competed with bacteria consortia added to the treatment, so that the TPH reducing bacteria can't work effectively. In any case, it can happen because there is competition between bacteria as stated by Yu et. al (2010), where an inhibitory effect will occur at the first week of treatment indicated by contaminant decreasing at a lower rate than the un-amended control soil.

Independent t-test explained that there were no significance in the comparison between any treatment for its removal efficiency. The correlation test also explained that there is a weak-inverse relationship correlation between bacteria population growth to TPH concentration change. The inverse relationship means, as the contaminant decreased, the bacteria population increased. This case is in accordance with the experiment by Inchor et al. (2014), where it was found that there is an inverse relationship between bacteria growth and the change of contaminant as a goods that bacteria utilize the contaminant's component for their needs. The determination coefficient was proof that only 9%-13% TPH removal was affected by bacteria population growth. It can be said that bacteria population growth is not the main cause of the decraesing value of THP. As stated by Tisma and Zeli (2010), fungi also have a capability to degrade TPH contaminant sometimes even better than the ability of TPH for reducing bacteria.

The table above explains the TPH removal rate, where the highest rate occurs for the treatment with 10% compost addition by 0,1016/day followed by treatment with 5% compost addition by 0,083/ day and tretment with 5% WWTP sludge addition by 0,0745/day, while the lowest comes from the treatment with 10% WWTP sludge addition by 0,0721/day. Based on this, we can conclude that it is not always the higher levels of nutrient that leads to bigger bacteria population growth. As already explained earlier, the composition of nutrient were also an important consideration to optimize bacteria ability in degrading the contaminant (Shahi, 2016). The result shows that at the end of simulation, 10% compost addition shows the highest percentage of TPH removal efficiency with 96,85% followed by soil amended with 5% compost which is 95,32%. While contaminated soil with 5% WWTP sludge residual presented 91,15% and 10% WWTP sludge residual addition presented 91,02% of TPH removal efficiency compared to un-amended control soil that showed 82,39% of TPH removal efficiency.

However, despite the highest TPH removal efficiency being reached in the addition of 10% compost, the fastest degradation occurs both in the addition of compost or WWTP sludge residual. Figure 7 shows that treatment with 5% compost addition and 5% WWTP sludge residual addition needs a longer time to reach the standard of 1% TPH concentration. Therefore, for this simulation, treatment with 10% compost addition has became the best treatment as it has the highest TPH removal efficiency and was the fastest to reach the policy standard of 1% TPH concentration.

#### **IV. CONCLUSION**

In conclusion, bioremediation can be an effective response to soil contamination by petroleum hydrocarbon. Biodegradation of soil contaminated by

Table 4 TPH removal rate.				
Treatment	TPH Removal Rate			
5% compost addition	0,9429			
10% compost addition	0,9753			
5% WWTP residual	0,971			
sludge addition				
10% WWTP residual sludge addition	0,9744			

#### Table 5 TPH removal efficiency

Treatment	Removal Efficiency
Control	82,39%
5% compost addition	95,32%
10% compost addition	96,85%
5% WWTP sludge addition	91,15%
10% WWTP sludge addition	91,02%

crude oil was positively enhanced by the amendment of compost and WWTP sludge.

The highest bacteria population reached log 10,06 for 5% WWTP sludge addition. Also the highest bacteria growth rate is 1,154 generation/week for 5% WWTP sludge addition. Independent t-test stated that the differences between all the treatments were not significantly different.

The percentage of biodegradation of soil recorded 95% (5% compost), 96,32% (10% compost), 91,15% (5% WWTP sludge), and 91,02% (10% WWTP sludge) higher biodegradation compared to control soil without nutritional amendments. The best biostimulant for this simulation is 10% compost addition.

#### SUGGESTIONS

- 1. Analyzed microorganism should be not only bacteria but also fungi or other microflora. Because fungi or microflora can have an ability to degrade TPH contaminant.
- 2. It is better to analyze nutrition availability periodically to know its utilization in the process.
- 3. Nutrition ratio should be analyzed to know which ratio will result in the best bioremediation process.

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